Quantum Dot Quantum Computing Program

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PROGRAM DESCRIPTION

The aim of this program is to develop a single photon source suitable for all-optical quantum computer technologies based on precise electrical control of a cavity quantum electrodynamical interaction between a single photon and a quantum dot (or other suitable "artificial atom"). The program is focussed on the use of high quality fused silica micro-cavities and associated fibre-based output coupling. Nanocrystal quantum dots are suitable for use with fused silica microcavities allowing strong quantum electrodynamical interactions between the cavity and nanocrystal, which can be used to carefully prepare and collect single photons with high efficiency. The various steps that are being undertaken are identification and characterisation of a quantum dot suitable for use in a single photon source, demonstration of the basic operational principles (such as an electric field induced Stark shift), manufacture and characterisation of sufficiently small fused silica microcavities and the demonstration and characterisation of an electrically controlled single photon source.

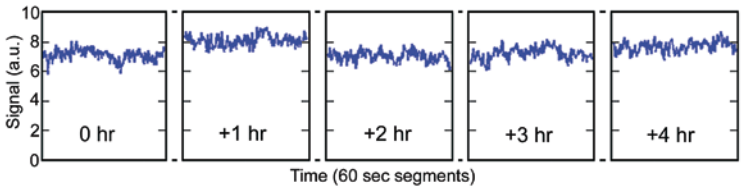

FIGURE 1

Evidence of blink-free emission and enhanced photophysical stability. A single nanocrystal emission trajectory obtained over five hours of steady blink-free emission. Each block represents a 1 min acquisition containing 200 points using a 300 ms integration time obtained at room temperature in vacuum. A pump irradiance of 1100 W/cm^2 is used, which is sufficient to drive multi-exciton states. There is no indication of blinking at this timescale.

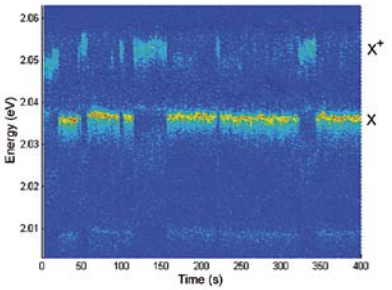

FIGURE 2

Spectral trajectory obtained from s single nanocrystal at 15 K with a pump irradiance of 1100 W/cm^2. Discrete jumps are observed between two different spectral positions. The jump energy of ~14 meV is found to be characteristic amongst a number of individual dots suggesting switching between an internal state. The switching characteristic, along with the energy separation, suggests that this state is a positively charged exciton or X^+ trion state. Thus we see considerable charge dynamics in single nanocrystal emission, even in the absence of observable blinking.

1. Dynamics of Non-Blinking Quantum Dots

Single nanocrystal photoluminescence is usually punctuated by periods of no emission, which occur suddenly in a binary on-off fashion. Such behaviour is referred to as blinking and is considered deleterious to many potential applications. This is especially true for the application as a single photon source. Furthermore, recovery from an "off" period usually leaves the nanocrystal in a different charge configuration state, which can also be problematic for a single photon source application. New CdSe/CdS/ZnS core/shell/shell nanocrystals developed in the Mulvaney lab have been studied at both cryogenic and room temperature and found to exhibit periods of non-blinking behaviour lasting for hours, even when illuminated at high intensities where multiphoton absorption dominates (Figure 1). Our spectroscopic study of these materials reveal that, while the nanocrystals do not enter an "off" state, they still display charge dynamics associated with the formation of and transitions between charged and neutral exciton states (Figure 2). These dynamics suggest that the charged exciton remains bright (as we have previously shown) in these materials. The implications of these findings are that new syntheses are improving the optical properties of these materials towards being more robust. Additionally, charged exciton states, which were previously considered to have a low photoluminescence quantum yield due to enhanced Auger recombination, can retain a significant photoluminescence quantum yield. Ultimately, charged exciton (or trion) states offer a very close approximation to a pure two-level system due to the exclusion of spin triplet states.

2. Sources of Spectral Instability

Single CdSe based nanocrystals are observed to exhibit significant spectral instability at the time scale common to our experiments (seconds to minutes). We sought to understand this instability and to characterize the time scale over which it occurs. Previously we had estimated the spectral instability timescale to be comparable to that of our experiment using a technique of high resolution Fabry-Perot spectroscopy. We have expanded on this work with the development of a statistical analysis of the spectral instability. When analysing long time series of spectral peak positions, we find that fluctuations in the spectral peak position are not completely random, but contain significant correlations. A theory that treats the analysis of the spectral jump data indicate that for Poisson distributed random jumps, a low jump rate will result in a strong correlation between successive jumps (Figure 3). The correlation is also found to be extremely sensitive to the integration period, as doubling the integration period removes the correlation entirely (Figure 4).

In general, the presence of the correlation reveals the spectral jump rate (during a single spectral acquisition period), N~1, from which we readily estimate the single nanocrystal spectral stability to be of the order of 10^4 emitted photons per single spectral jump. Such a low jump rate reveals that the spectral instability is only weakly coupled to the initial photo-generated exciton.

However, further analysis of the data indicates that there is an additional contribution to the observed correlation. We find that the observed jump distribution is independent of the integration time, while the correlation is strongly dependent on the integration time. These properties cannot be reproduced using purely random jump models, suggesting that the jumps are also correlated. We indeed find an anti-correlation between successive jumps when we analyse the jump values, rather than their absolute values. Such correlations are predicted when considering fluctuating two-level-systems with highly asymmetric excitation/relaxation times. Once more, the correlations are predicted only for jump rates, N~1, compatible with our previous analysis, but are more sensitive to changes in the jump rate, N.

An important outcome of this work is that the dominant spectral instability is identified as a single physical process. This rules out random environmental charge motion and

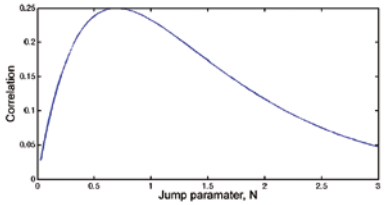

FIGURE 3

A plot of the theoretical jump rate dependence of the correlation that can be observed with Poissonian statistics for low jump rates, N.

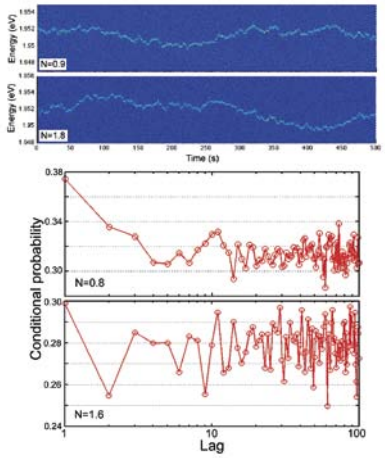

FIGURE 4

Simulations of correlations in spectral diffusion. Two sets of simulated single nanocrystal spectra are shown, generated using a random spectral jump rates of N=0.9 and N=1.8. We note that the two spectral traces show no evidence of the difference in jump rates. When we plot the conditional probability that a second spectral jump resides in the first spectral bin, *I* jumps after finding the first jump resides in this bin ($P_i(\nu \in b_i \,|\nu \in b_i)$, where b_1: $0 \leq \nu < 60$ μeV), we find that there is a pronounced correlation spike in the N=0.9 data, which is significantly reduced in the N=1.8 data, illustrating the sensitive dependence on the jump rate, in agreement with our observations.

points to a surface ligand instability as a likely cause. It is most likely that the process corresponds to the photo-induced removal and re-attachment of single ligand molecules from the surface of the nanocrystal. This identification should facilitate means of improving the nanocrystal synthesis in order to minimise or remove this instability entirely through the appropriate choice of surface ligand

3. Surface Effects on Exciton Spin Lifetime

We have found that the luminescence from single CdSe/CdS/ZnS core/shell/shell nanocrystals usually show spectral doublets, which we have shown to be emission from both the bright and dark exciton fine structure states. The observation of spectral doublets thus represents a significant improvement in the photo-physical properties of these materials, as they indicate that an efficient spin relaxation pathway can be controlled using synthetic procedures. We have quantified the spin relaxation rate in different single nanocrystals using thermal redistribution of population in conjunction with a rate equation analysis. Spin lifetimes longer the bright exciton lifetime are found, which are now comparable to those found in self-assembled quantum dots comprising state-of-the-art single photon sources. Our results also indicate that this spin relaxation channel can be manipulated using different surface treatments. New materials based on variants of the CdSe/CdS/ZnS core/shell/shell structure have exhibited remarkable photo-physical stability with high brightness and many hours of blink-free emission. However, these new materials are also remarkable in the near complete absence of spectral doublets. In contrast, materials previously obtained from the same lab were found to exhibit a high proportion of spectral doublets. This discrepancy seems to be related to the surface treatment, as the core/shell/shell structures are similar in all cases. Thus we are poised to uncover the role of the surface in mediating the exciton spin relaxation in these materials, with the aim of suppressing the spin relaxation mechanism.

4. Coupling of Quantum Dots to Micro-Cavities

We have started investigating nanocrystal quantum dots coupled to high quality micro-toroids supplied through our collaboration with Dr Warwick Bowen. These micro-toroids represent the state-of-the-art in high-Q microcavities and offer the advantage of rigid attachment to a silicon substrate, enabling convenient input/output coupling via a fibre taper. Initial experiments were conducted at room temperature with nanocrystals deposited on the micro-toroids using a micropipette to place a 10 μl droplet of colloid solution on each toroid. While single toroids could be targeted, surface tension and wicking tended to spread the solution over as many as five toroids in the linear array. Imaging of the toroids was undertaken in the fluorescence microscope and single nanocrystal emission was easily identified (Figure 5). A range of different doping concentrations was employed, from a low concentration, where single isolated nanocrystals were observed, to higher concentrations leading to a much higher density of emitters and a more uniform coating. Spectra of the dots on the microcavities were gathered with the intention of detecting coupling to cavity modes, as sharp dips in the emission spectrum. However, no evidence for coupling to the cavity modes has yet been found. This is believed to be a consequence of the high cavity-Q of the resonances combined with the relatively low spectral resolution of the apparatus.

Micro-toroids doped with quantum dots were also investigated by coupling light into the micro-toroid using an adiabatic fibre taper. A scanning diode laser then probes the cavity modes. In this case the diode laser

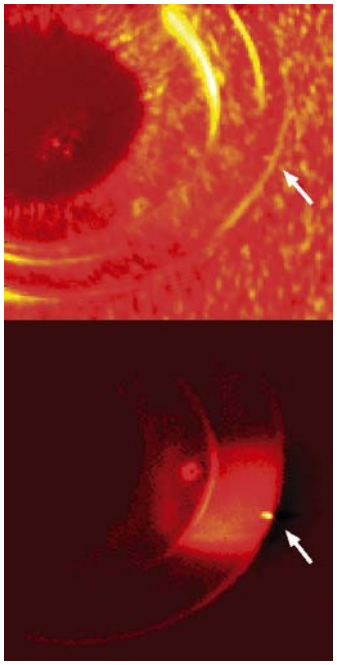

FIGURE 5

Images of single nanocrystal emission on a micro-toroid cavity.The white arrows mark point to the single nanocrystal emission. The top picture is an image using LED illumination in order to image the toroid. The lower picture is purely a luminescence image, using 532 nm laser wide field excitation.

operated at a wavelength far below the band-gap of the nanocrystals, where there is negligible absorption. Coupling of the nanocrystals to the cavity modes was confirmed as an increase in intra-cavity back scattering, which results in a spectral doublet due to the coupling between the forward and back-scattered fields. The increased coupling between the forwards and back-scattered fields indicates that the nanocrystals are indeed coupled to the intra-cavity field.

An experiment aimed at studying the cavity-enhanced two-photon interaction is underway. Cavity-enhanced two-photon absorption using nanocrystals has the potential to generate non-classical states of light and ultimately reduce a weak coherent state into a train of indistinguishable single photons, with the advantage of operating at room temperature. Furthermore, the room temperature study will also pave the way for the cryogenic experiment, which will be built in a new Helium bath cryostat due to arrive in 2010. The apparatus consists of a 3-axis piezo-stage, two long distance imaging microscopes and a fibre-taper holder for aligning the micro-toroid with the fibre-taper. A scanning external cavity 780 nm diode laser is being used to probe the system, with the intention of pumping a two-photon absorption in the nanocrystal/micro-toroid system. The output from the toroid will be detected using the current fluorescent microscope, which is equipped with the appropriate filters.